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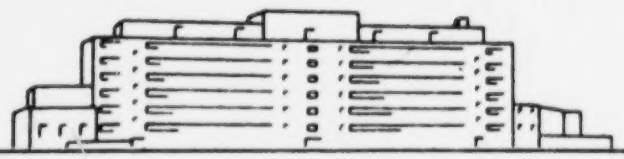
PRELIMINARY APPRAISAL OF ADVANCED WASTE TREATMENT PROCESSES



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Public Health Service

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PRELIMINARY APPRAISAL OF ADVANCED WASTE TREATMENT PROCESSES

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U. S. DEPARTMENT OF HEALTH, EDUCATION, AND WELFARE

Public Health Service

Robert A. Taft Sanitary Engineering Center

Cincinnati, Ohio

September, 1962

EDITORIAL NOTE

This report, the second in a series being published to present significant findings achieved in water renovation under the Advanced Waste Treatment Research Program, was submitted in fulfillment of a contract (Purchase Order No. 3159) between Harvard University and the Public Health Service. Although over-all guidance of the project was provided by the AWTR Program, the results obtained, opinions expressed, and conclusions reached are those of the authors and are not necessarily endorsed by the Department of Health, Education, and Welfare. Except for editorial review, this report appears as received from the contractor.

CENTER PUBLICATIONS

The Robert A. Taft Sanitary Engineering Center is a national laboratory of the Public Health Service for research, training, and technical consultation in problems of water and waste treatment, milk and food safety, air pollution control, and radiological health.

ADVANCED WASTE TREATMENT RESEARCH PROGRAM

The Advanced Waste Treatment Research Program of the Public Health Service has two ultimate goals: One is to help abate out Nation's growing water pollution problems and the other, more startling in concept, is to renovate waste water for direct and deliberate reuse.

Today's water and waste treatment processes, and we must include natural purification by our rivers and streams, are not capable of removing all contaminants from water. The resistant contaminants, or materials, range from simple inorganic salts up through an ever-increasing number of highly complex synthetic organic chemicals, and are termed "refractory" materials. To develop a new technology for removal of these materials from water is mandatory if we are to protect the quality of our Nation's water for the entire range of human and industrial needs.

Beyond this, it must be recognized that waste waters constitute this country's most immediately available untapped water resource. They are available at every location at which water is used. They do not have to be pumped long distances, over mountains, or from deep underground sources. When we learn how to renovate these waters efficiently and economically, a new dimension in water resource management will be opened.

In one step, then, advanced waste treatment, i.e., water renovation, could alleviate both water pollution and water supply problems--problems of increasing concern both in this country and in the rest of the world. The importance of advanced waste treatment was recognized by the Congress in its 1961 amendments to the Federal Water Pollution Control Act (Public Law 87-88). This act directs the Secretary of Health, Education, and Welfare "...to develop and demonstrate practicable means of treating municipal sewage and other water-borne wastes to remove the maximum possible amounts of physical, chemical, and biological pollutants in order to restore and maintain the maximum amount of the Nation's water at a quality suitable for reuse."

Technically, advanced waste treatment may be looked upon as a two-step process: (1) separating concentrated contaminants from the purified water "product" and (2) disposing of the concentrated contaminants in a way that will render them forever innocuous. This "permanent disposal" of separated wastes is an important part of the advanced waste treatment concept. If contaminants are discarded back into useable surface or ground water supplies, pollution problems will not be diminished and downstream water users will have to remove the same contaminants again and again.

The need for developing an advanced waste treatment technology is far from academic. It is clear today that we are using

and reusing our fresh water supply over and over. Four persons in ten in the United States now use water that has already been used at least once for domestic or industrial purposes. In many areas, multiple reuse exists. It is a hard fact that our inexorably increasing water reuse needs will inevitably overtax the capacity of streams and water treatment plants to remove potentially damaging contaminants from our water supplies unless improved purification processes are developed and brought to use. Water quality has been damaged in some areas, as exhibited by foaming, fish kills, taste and odor problems, and impairment of municipal and industrial water treatment processes. Concern is emerging over possible long-term effects on human health.

The challenge, the measure of success, in advanced waste treatment research is economics. Complete water renovation is already technically possible, but at unreasonably high cost. In addition, tomorrow's economics must apply rather than today's. The public now spends an average of about 23 cents per 1000 gallons in direct costs for a single water-use cycle, exclusive of water distribution and waste collection expense. In prevailing high-cost areas, this figure is at least 30 cents per 1000 gallons. A water renovation system would perform the functions of this cycle, and perform them in a manner adequate to meet the needs of the future. Chemical, biological, and radiological pollutants would be removed from our water courses; taste- and odor-bearing materials and potentially toxic substances would be absent from our tap waters; the economic growth of whole regions would not be hindered by problems of water supply.

It is too early to predict the cost of advanced waste treatment technology, but the need is inevitable. Because of population increase and industrial expansion, we are approaching the time when our rivers and streams will no longer be able to assimilate our wastes and when there will be no more developable supplies of fresh water. Whatever the cost of this technology, one thing is certain--it must be the very lowest science and engineering can achieve. Any thing more will impose an inexcusable, perhaps intolerable burden on our country's economic growth and on its ability to compete in tomorrow's world.

FOREWORD

Early in the development of the Advanced Waste Treatment Research Program, the expert counsel of Professor J. Carrell Morris of Harvard University was sought. Specifically, Professor Morris was requested to make a preliminary appraisal of waste water reuse and ways and means of economically removing the contaminants in waste water that are not removed by today's best conventional sewage treatment practices. The processes to be considered were to include, but were not limited to, adsorption, flotation, chemical oxidation, and chemical coagulation. The contractor had free choice as to other processes to be discussed.

Professor Morris enlisted the able assistance of Dr. Walter J. Weber, Jr., who at the time of writing the report was a doctoral candidate at Harvard University.

Both technical and brief economic evaluations of the processes considered are included in this final report of their study. Adsorption, oxidation, and coagulation as well as processes for the removal of specific pollutants are discussed in detail. The authors present an excellent bibliography on these processes and on recorded treatment and uses of waste waters for various purposes.

This report represents the results of the authors personal experience and knowledge and a thorough literature search. It is a preliminary appraisal of the problem of removing refractory materials from waste water and is published as part of the record of the Advanced Waste Treatment Research Program.

F. M. Middleton, Chief
Advanced Waste Treatment
Research Program

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PRELIMINARY APPRAISAL OF ADVANCED WASTE TREATMENT PROCESSES

GENERAL CONSIDERATIONS

This report provides a preliminary appraisal of currently proposed means and methods for economically securing more complete treatment of waste waters than is customarily attained. The appraisal is based upon a careful consideration of ideas and proposals set forth in the recent literature in the water supply and waste treatment fields.

In the wake of a rapid population growth, the nation, and indeed the world, is presented with the acute problem of increased pollutional loads on streams, rivers, and other receiving bodies. From this has developed a well-defined need for more complete treatment of waste waters than is achieved by conventional methods, particularly in view of the increasing need for water reuse. Few of those who are cognizant of the present water situation will question the need for more stringent purification of waste waters in treatment plants prior to discharge of the waters into receiving bodies. The mushrooming population has resulted in the development of an urbanized society, enveloping its own life-sustaining watercourses. As a consequence, the waste discharge by one urban area into a receiving stream or river may well serve as a water supply for another, with little or no natural purification between expulsion and subsequent intake. On occasion the discharging area may well be utilizing, knowingly or unknowingly, a part of the discharge for its own water supply.

Coupled with the increase in pollution as a result of population expansion is the problem of the increased use and disposal of substances that are not readily broken down biologically. In this category are alkylbenzenesulfonates and other synthetic detergents, pesticides, and polymeric materials.

Approximately 65 percent of the population of the United States now enjoys the benefit of public water supply, and even today nearly 60 percent of this group is supplied with water that has already been used at least once for industrial or domestic purposes.[1] At a minimum, then, nearly 40 percent of our entire population is using water that in part has previously been returned to the source as a sewage effluent. The need for more complete treatment of waste water becomes obvious, and immediate. As Hol-

lis[2] has pointed out, under existing reuse conditions and those expected in the years ahead, the concept of the assimilation of pollution by streams and other receiving waters completely breaks down as a major usable factor in waste treatment practices.

WATER REUSE

The use, and reuse, of water may be divided into three main categories, in order of increasing usage: domestic, agricultural, and industrial. Merz[3] has referred to four classes of waste water utilization: industrial, agricultural, recreational, and ground water injection. The last class pertains partially to each of the three major groups given in this report; the recreational class could be included in domestic usage.

Rawn and Bowerman[4] have noted several plants currently in operation that are designed to provide effluents specifically intended for irrigation or for industrial purposes. The principal deterrent to the direct use of a domestic sewage as a raw water source is its unsuitability from an esthetic point of view. There is, however, as has been previously pointed out, much indirect use of sewage effluent as a raw water source.

It has been estimated that approximately 25 percent, or 50 mgd, of the raw sewage of the Los Angeles County Sanitation District can be economically reclaimed.[4] Howe[5] has described a waste treatment system at Eli Lilly and Co. that produces a highly purified effluent from 1.05 mgd of strong wastes from processes for the production of antibiotics. Some indication of the economic feasibility of the reclamation and reuse of sewage plant effluents is given by Levy and Calise.[6] These authors have noted that many plants, particularly in Europe, are already using sewage effluents to good advantage and have furnished economic and analytical data to substantiate this.

Domestic Application

An example of the reuse of waste waters for domestic purposes has been cited by Laverty.[7] The Los Angeles County Flood Control District embarked on a study of the feasibility of using sewage plant effluent for recharging ground water supplies, in an effort to combat sea-water intrusion. Preliminary data indicated that the Hyperion Treatment Plant effluent, in which suspended solids and BOD varied from 30 to 80 milligrams per liter, could be injected into the ground water system without resulting in a violation of established drinking water standards.

The British Water Pollution Research Laboratory has also studied the recharging of ground waters with sewage effluent.[8] The changes produced when polluted water percolated through 2 feet of porous Butner sandstone were observed in these studies. Intermittent application of sewage plant effluent at a rate of 0.3 foot per hour for 12 hours each day substantially reduced ammonia-nitrogen and BOD at both 9° and 15°C. The results indicated that ammonia-nitrogen is retained in the soil by an exchange process and is leached out after it is oxidized to nitrate. Data were presented to support this viewpoint. The report of the study also included information on changes in dissolved oxygen and in bacterial densities.

Metzler et al.[9] have described a dramatic instance of waste water reuse for domestic purposes. Chanute, Kansas, during the severe midfifties drought, cycled treated sewage through a stabilization pond, the water treatment plant, and the water distribution system. The mean cycle time of the water moving through this temporary emergency system was only 20 days.

For 2 of the 5 months that the recycling system was employed, the only special precaution provided was the recirculation of the sewage in the treatment plant. For the remaining 3 months extensive chlorination was practiced so that the bacteriological quality of the tap water was always maintained within the Drinking Water Standards. The concentration of parasitic protozoa in Chanute's sewage was low during this period, and these organisms were not found in the treated water. The cysts of free-living amoebas were found regularly in the treated water, however, indicating that the treatment plant had a relatively low efficiency for removal of such organisms. No adverse effects on the health of the people of Chanute were reported, although viruses of the poliomyelitis, ECHO, and Coxsackie groups were isolated and identified in treated sewage samples. No virus was detected in the treated water, however.

The water at Chanute was reused approximately 8 to 15 times.[10] The treated water gradually developed a pale yellow color and an unpleasant taste and odor. It contained undesirable quantities of dissolved minerals and organic substances, and foamed readily when agitated. Chloride increased about 32 milligrams per liter for each use, or from about 40 to 670 milligrams per liter over all. At its worst the raw water contained 17 milligrams of combined nitrogen, 5.8 milligrams of alkylbenzenesulfonate, 670 milligrams of chloride, 13 milligrams of BOD, and 1000 to 1200 milligrams of total solids per liter.

The character of the raw water resulted in the following conditions at the water treatment plant: (1) inability

to practice free residual chlorination for taste and odor control and color removal because of the extremely high chlorine demand of the raw water, (2) frothing at the recarbonation basin and in the rapid sand filter during backwashing, (3) less effective coagulation and settling, (4) plugging and coating of the filter sand, and (5) false high readings of free chlorine residuals obtained by the OTA test.

It should be noted that the reuse of the water practiced in Chanute far exceeded the reuse limits suggested by Imhoff[11] many years ago. Imhoff presented a general discussion of the possible cycle limit that sewage can be exposed to if it is still to be economically treated to give a potable water. He concluded that approximately nine cycles is the limit, based mainly on salinity buildup.

The use of sewage effluents for supplying domestic water requirements is the ultimate in the reuse of waters. It has been demonstrated that this is possible, though not necessarily desirable as presently practiced. The real advantages of the reuse of waters may be better realized in the agricultural and industrial areas.

Agricultural Application

Goudey[12] was one of many to realize the potential of sewage effluents for the irrigation of desert lands in the California area. Some areas in the United States with low annual rainfall are currently using sewage effluents for irrigation purposes. This is particularly true of the livestock feed and pasture crops in the Western section of the country, e.g., Washington, California, Arizona, Nevada, Texas, etc. Crops irrigated with sewage effluents include corn, hay, grain, walnuts, fruit, alfalfa, maize seed, cotton, potatoes, orchards, beets, beans, peppers, and so on.

It is common practice not to use any form of sewage on crops that are to be eaten raw. Sewage-borne organisms have been found in marketed vegetables and fruits on which sewage effluents were used for irrigation of the crop. It has been reported[1] on the basis of recent experiments in Germany that sewage-borne organisms are still contained in potato tubers 40 days after application of sewage, in carrots 10 days, and in cabbage and certain berries 5 days.

An outstanding example of the large-scale reuse of water for agricultural purposes is current practice in the state of Israel. Municipal sewage is used in Israel for growing feed crops, stabilizing shifting sands, and raising the water table. The true benefit of using sewage effluent for irrigation in Israel, or anywhere for that matter, may not lie only in its water value but also in its nutrient content.

Merz[3] cites three benefits to a community using domestic sewage effluent for agricultural purposes: (1) a safe satisfactory disposal for waters that might otherwise cause stream pollution; (2) a conservation of other waters for more beneficial use; and (3) agricultural nutrients for the land.

Stone and Merrell[13] have given a detailed report of the significance of minerals in waste waters, as they relate to agricultural reuse. They have summarized the results of field studies and letter surveys of 25 sewage systems, showing the major mineral components of sewage effluents and comparing these with the United States Department of Agriculture standards for irrigation water.

Industrial Application

Treated sewage effluents have their greatest potential for reuse in industrial processes. Today only an estimated 1 percent of the total municipal wastes of the country is being utilized directly by industry, but this small utilization is so successful that the author has predicted a much fuller realization of potential within a very few years.

Keating and Calise[14, 15] discuss conditions necessary for the industrial reuse of sewage effluents and the effects of various characteristics of sewage effluents on industrial reuse. Some of the obvious conditions include a demand for water for processes that do not affect public health, a volume of effluent that is reliably large enough, an effluent that is consistently suitable in quality, and processing costs that are less than those for an alternative supply. The characteristics of sewage effluents that are discussed by Keating and Calise in connection with their effects on industrial reuse include color, concentration of nitrogenous compounds, BOD concentration of dissolved solids, and the presence of phosphates.

Several recent publications[16-22] cite specific examples of industrial reuse of sewage effluents. Case histories of nine reported instances of such reuse have been given by Veatch.[23]

Amarillo, Texas, was able to finance the construction of a new sewage treatment plant through a contract with a neighboring oil refinery for the purchase of effluent from the proposed treatment plant for use in cooling towers, boilers, etc.[24] The cost of the reclaimed effluent has been reported to vary between 11 and 17 cents per 1000 gallons, as compared to 18 cents per 1000 gallons for fresh water.

It has been estimated[1] that 50 percent of all industrial water is used for cooling and boiler feed. This represents an extremely valuable potential application for the reuse of

sewage effluents, and provides a possibility of preserving tremendous volumes of fresh water for domestic use.

Water Reuse Deterrents

The prime deterrent to the reuse of secondary effluents and the most critical factor related to pollution of receiving water are the buildup of organic compounds that are not readily degraded biologically in the effluents or subsequently in the receiving waters.

The number and variety of exotic organic materials that find their way into streams, rivers, and lakes are rapidly increasing. For example, insecticides and pesticides, such as dichlorodiphenyltrichloroethane, dichlorodiphenyldichloroethane, and other chlorinated hydrocarbons, are often used indiscriminately in agricultural and related applications and by one course or another reach streams and rivers in rather high concentrations. They are introduced into sewage by infiltration from surface drainage and in wastes of processing industries. When totally resistant to biological degradation, these materials usually escape in the effluent from the treatment plant in much the same concentrations as they were present in the influent. With successive reuse such substances build up to levels that may be harmful to subsequent consumers of the effluent, to say nothing of toxic effects on fish and wildlife living in or about the receiving waters.

Detergent compounds, especially alkylbenzenesulfonates, constitute an even greater problem. Certain types of synthetic detergents have been clearly identified as one component of raw sewage that strongly resists conventional treatment. Such materials are finding ever-wider application in household and industry, and they present a problem that must be dealt with immediately if grossly obnoxious pollution of receiving waters is to be avoided.

There is, in addition, a long list of other organic compounds that originate in industrial processes and are eventually discharged into watercourses. Further complicating the problem is the great number of interactions possible between the various compounds, resulting in formation of completely unsuspected organic materials.

Much difficulty has been encountered in the identification and enumeration of the soluble organic constituents of secondary effluents. Studies to date have left nearly 60 to 70 percent of all soluble organic compounds in sewage effluents unidentified. Detergents comprise the major portion of those compounds that have been identified.

As a result of this situation practical methods for the further purification of treated sewage must be of such a

nature that they can remove a broad variety of organic materials of urknown constitution. Consequently, approaches to the problem up to now have been almost purely empirical and have provided little generally applicable information on removal efficiencies, selectivities of removal, costs of operation, and the like.

PROCESS CONSIDERATIONS

The value of more complete treatment of waste waters is well recognized. The major problem is the development of means for economically achieving a reusable sewage effluent. More complete treatment than the usual primary sedimentation and secondary biological oxidation has been referred to as "tertiary" treatment. This is perhaps somewhat of a misnomer in that a much more completely treated effluent may be achieved by improving conventional first and second stages of treatment, or perhaps by a newly devised method in the complete absence of the so-called "primary" and "secondary" stages. In this review, however, the term will serve, since discussion will deal chiefly with a third stage of treatment imposed upon the effluent from a conventional secondary treatment plant.

The first stage of waste water treatment is primarily a physical process of screening and sedimentation, and the second stage is biological. Some form of chemical treatment seems indicated for the proposed third stage. It should, of course, be economical and possess relative ease of operation. Consequently, it appears that such a process should be a relatively simple chemical operation involving inexpensive reagents. No truly significant advances in methods for waste treatment have been made in the past few decades. It is true that many modifications have been introduced, but these methods, as they are presently known and practiced, seem to offer no real value as operations for more complete treatment. Secondary effluents have supposedly undergone biological treatment; thus, the nature and concentration of remaining impurities are such that further biological processing does not present attractive possibilities. The most promising types of operations are, in order of prospective merit:

1. Adsorption.
2. Coagulation.
3. Chemical Oxidation.

Chemical oxidation appears likely to be quite expensive for achieving the desired degree of purification. Processes based on coagulation and adsorption are somewhat similar, since removal of impurities by coagulation is in part a result of adsorptive action. The distinction is that when coagulation is used the adsorbent is formed in the medium rather than being added in a preformed condition.

A comparative evaluation of coagulation, adsorption, and chemical oxidation methods for the purification of waste water containing byproducts from the production of benzidine and aromatic sulfonic acids has been carried out by Sterbacek and co-workers.[25] They have reported that the best and most economical results were achieved by adsorption on activated carbon preceded by coagulation with aluminum sulfate.

Adsorptive Processes

As mentioned, a study of the literature has indicated that use of adsorbents and adsorptive processes is the most promising of currently known methods for the further treatment of secondary sewage effluents. Research in this area has been initiated at Harvard University and by several investigators at other institutions.

Most of the literature on this subject deals with the removal of specific impurities from wastes. Typical substances reported as having been removed by adsorptive processes are: synthetic dyes,[26, 27] acids,[26, 28] salts,[26, 28] phenols,[28-38] insecticides,[28, 50] fungicides,[28, 50] turbidity,[39] TNT,[40] H_2S,[27] SO_3,[27] S_2O_3,[27] alkylbenzenesulfonate,[28, 41-43] butadiene,[44] ethylene chloride,[44] tetrachloroethane,[44] trichloroethylene,[44] cresols,[28] nitro compounds,[28] benzidine,[25] and humic material.[45] This incomplete list shows the variety of materials subject to removal by adsorption and indicates the potential of the method.

Great variety exists, too, in the type of adsorbent used. For example, Renn and Barada[41] have reported laboratory studies dealing with the adsorption of alkylbenzenesulfonate on soil, silt, china clay, diatomite, silica gel, paraffin, calcium carbonate, and activated carbon.

Activated carbon is presently the most widely used of all adsorbents. Sigworth[46] has discussed factors influencing the efficiency of adsorption on activated carbon. Many of these factors apply to any or all adsorbents and may be summarized as follows:

1. Amount of carbon applied. This varies from 1 to 1000 milligrams per liter in extreme cases, the average being 3 or 4.
2. Degree of wetting. For best adsorption the carbon must be thoroughly wetted.
3. Degree of dispersion. Efficient adsorption requires adequate dispersion of the carbon in the fluid. This can be accomplished by applying the carbon at pump suctions, mixing basins, and so on.
4. Continuity of application. A fairly uniform level of removal should be maintained.

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5. Contact time. Approximately 15 to 20 minutes is needed for maximum adsorption.

6. pH. Adsorption is best when the water is slightly acid or neutral, although carbon is effective up to pH 9.0. It is advisable not to mix carbon with alkaline compounds, and it should be applied before lime or soda in softening plants in order to avoid a loss in adsorption efficiency at high pH.

7. Temperature. Low temperatures are slightly more favorable for adsorption than high temperatures.

8. Chlorine application. Activated carbon acts to dechlorinate water; hence, it is advisable to apply carbon before chlorine. An additional advantage of this order of application is the reduction in chlorine-demanding materials.

Carbon has been used as an adsorbent in various physical forms. Dierichs and Jaehn[29, 32] have reported the adsorption of catechol and its homologues on powdered carbon. They indicate that there is a catalytic effect of the powder on the conversion of the phenolic materials to water-insoluble polymers. Other investigators have found beds or towers of carbon, through which the water is allowed to percolate, to be very effective.[27, 28, 31] Kul'skii et al.[34] have reported the use of a series of clarifiers containing a suspension of powdered activated carbon stabilized by the addition of 10 percent aluminum oxide. Draper has recently discussed properties and uses of various types of activated carbon.[51]

Clay has been used successfully as an adsorbent in the removal of dyes[26] and humic materials.[45] Van Beneden has stated that water colored by humic materials may be decolorized by adsorption of the humic material on clay particles.[45] It can be said that under such circumstances the humic particles most likely peptize the clay.

Fly ash appears to be a very promising adsorbent; however, the chemical and physical characteristics of the materials meant by this designation have generally not been reported. Freston[47] has presented a general discussion of the sources and uses of fly ashes and slags. Solin and Kustka[40] have used generator and furnace ashes for the adsorption of TNT at various pH values. The ash was regenerated by aeration. The generator ash proved somewhat more efficient, adsorbing 0.801 percent its own weight in TNT from a solution containing 100 milligrams of TNT per liter at pH 6.8. The furnace ash adsorbed 0.754 percent under identical conditions. Used in a filter or column, the ash removed 90 percent of the TNT from the solution passed over it in the course of a year. During this period, the weight of TNT passed over the filter beds was over 100 times the weight of the ash.

Rieche and Strankmuller[30] have described a process by which phenols are removed from an industrial waste in order to facilitate further biological treatment. Air is blown through the waste to which slag has been added. As the phenols are oxidized, the slightly acidic products of oxidation are adsorbed tightly to the slag. A very similar process has been reported by Brabek.[33, 38]

Lorenz[37] successfully employed fly ash and grate ash in the final purification and dephenolation of a phenol-containing waste water. Both simple and complex phenols were reported to be effectively adsorbed.

The economical regeneration of adsorbents has received a good deal of attention. The reactivation of carbon is of great importance because of its relatively high initial cost. Among the many methods that have been attempted are the use of superheated steam, sodium hydroxide, and hydrochloric acid.[48] Johswich[49] has presented a discussion of the economic aspects of regenerating activated carbon to as much as 70 to 90 percent of its original activity.

There yet remains to be found a simple, inexpensive adsorbent that may be regenerated with ease or that may be discarded completely after one use. If and when this is done, it seems likely that adsorptive processes will be left nearly unchallenged as the logical choice for the further treatment of sewage effluents.

Chemical Coagulation Processes

It is not at all difficult to understand the interest in chemical coagulation as a possible means for attaining a higher degree of purification of waste waters. For years this process has served as one of the most widely used and most versatile treatment methods available for removal of unwanted substances from water supplies. Much recent work has been done in exploring the many facets of chemical coagulation and in defining significant parameters.

Classical concepts of the mechanics of the sedimentation of flocs are being re-examined and challenged. Research in the fields of sanitary engineering and industrial processing is throwing new light on basic principles, and coagulation and subsequent sedimentation are becoming more science than art. Budd[52] has stated that research at Dorr-Oliver led to the delineation of four markedly different types of settling: (1) clarification of dilute suspensions of discrete particles, governed by detention time and overflow rate; (2) clarification of flocculent material, governed by detention time and overflow rate; (3) zone settling of coherent solids; and (4) compression of flocculated particles at the bottom of a basin, governed by solids detention time and pulp depth.

Stones[53] has discussed laboratory investigations on the changes taking place during the settlement of a flocculated sewage. The study referred to was limited to laboratory-scale quiescent settling in order to avoid the great diversity of factors involved in tank treatment and to decrease the possibility of secondary effects beclouding the fundamental considerations. The studies showed that the changes occurring in the settlement of the floc are greatly influenced by biological activity, and it was pointed out that, in the continuous flow of sewage through tanks, Hazen's theory of sedimentation does not nearly account for the degree of purification attained.

If the process is so dependent on biological activity, it follows directly that the temperature becomes an important parameter, as does, for another example, the presence or absence of toxic material.

Stones[53] has recommended that chemical precipitation to remove finely divided suspended solids and colloids should not be practiced indiscriminately. Studies showed that with long detention times small dosages of lime or acid may actually hinder the degree of purification. Stones also discussed the effects of the iron, chromium, and phosphate contents of waste waters on chemical coagulation and sedimentation.

In a patent review, Barham, Matherne, and Keller[54] have surveyed the various devices proposed and developed in the past 106 years for the separation of finely divided suspensions into relatively clear effluents and thickened sediments or sludges. The three phases of flocculating, settling, and thickening as carried out in clarifiers, settling tanks, separators, and thickening tanks were covered.

Induced vibrations are known to have a markedly beneficial effect on the sedimentation of some suspended matters. However, there has not been much written on the application of this device to waste water treatment. Lyon[55] discussed it to some extent in an article written some years ago.

The use of organic coagulants and coagulation aids is becoming common. Organic flocculants having cationic groups are economically attractive substitutes for inorganic coagulants in many applications.[56] An example that has been reported by Clemens[57] is the purification of foundry sand wash water by use of hydrophobic polyamines. The use of inorganic coagulants such as aluminum sulfate in conjunction with organic aids may provide a more useful approach to the problem of chemical coagulation of waste water. Although in many instances the use of organic coagulants, such as Separan 2610,[58] has been found to give excellent results, the cost has been considerably greater than with conventional coagulants.

Cohen, Rourke, and Woodward[59] have reported studies with anionic, cationic, and nonionic coagulant aids, and have given a detailed evaluation of natural and synthetic polyelectrolytes. Acrylic acid derivatives have been used with good success on a fairly large scale in the clarification of coal-washing waters.[60, 61] Water-soluble cellulose derivatives and alkali-soluble, water-insoluble derivatives have proved very satisfactory as coagulant aids when employed in concentrations from 0.1 to 10 milligrams per liter.[62]

Aluminum sulfate is still the most widely used coagulant, and a rather good summary of the applications of alum, along with removals achieved, has been presented by Harwood.[63] Sodium aluminate has also been used as a coagulant.[64] The ash from some coals, activated with sulfuric acid, has been reported to be superior in many instances to aluminum sulfate in the treatment of waste waters, because of its high sorptive power, high rate of sedimentation, and low cost.[65] The reclamation and reuse of ferric sulfate coagulant used in treating oil emulsion wastes has been reported as economically feasible in specific instances.[66] Inorganic ferrates, perferably alkali metal ferrates, have been found useful in coagulating wastes containing cyanides, phenols, and chlorophenols, since they serve the additional purpose of converting the carbon of these organic constituents into carbon dioxide, thereby destroying the organic compound.[67]

Much of the literature necessarily deals with the coagulation of specific wastes or specific substances. These substances or wastes include: phenols,[68, 69] fluorides,[70, 71] paint suspensions,[72] chromate wastes,[72] TNT,[73] radioactive substances,[74, 75] iron compounds,[76-79] manganese compounds,[76, 77, 79, 80] laundry wastes,[81] color,[80, 82] odor,[80] pharmaceutical wastes,[83] sulfur-containing mine wastes,[84] phosphates,[85, 86] dredge-mining wastes,[58] oils,[69, 87-89] paper and pulp mill wastes,[90] and synthetic detergents.[91]

An interesting description of a coagulating process for the rapid clarification of paper mill waters has been given by Stemen.[92] First, the waste water is rapidly mixed with a coagulating chemical and a ferromagnetic powder. Second, the resulting mixture is gently agitated to promote the formation of a floc containing both waste impurities and the ferromagnetic powder. Third, the floc is rapidly removed from solution by a magnetic drum over which the solution is passed. This final step involves only a few seconds.

The main problem with the use of coagulation in the further treatment of sewage effluents is its relative ineffectiveness, or lack of knowledge concerning its effectiveness, in the removal of soluble impurities. For example, alkyl-

benzenesulfonates are not adequately adsorbed by standard coagulants as they are now used.[59] However, the operation of the process of coagulation is so simple and economic factors appear so attractive that further investigations of modified processes or of new coagulating substances seems warranted.

Chemical Oxidation Processes

A third possible type of treatment for sewage effluents is the use of oxidants. Oxygen, ozone, chlorine, and chlorine dioxide have been tried as oxidizing agents in waste water treatment. The same types of considerations apply to the use of any oxidizing agent that can be efficiently and economically obtained and employed.

The simplest source of an oxidant is, of course, the atmosphere, but this rarely proves to be an efficient source. One reported instance of the use of atmospheric oxygen is for the oxidation of phenols in a municipal sewage.[33] Generally, the use of atmospheric oxygen has proven impractical, however, because of slow oxidizing action at ordinary temperatures.

Aeration under pressure has been used with fair success in the oxidation of wastes from chemical operations,[93] wastes containing byproducts from the production of benzidine and aromatic sulfonic acids,[25] and paper and pulp mill wastes.[90] However, the most promising use of compressed air or oxygen seems to be for the oxidation of phenols,[28, 30] cresols,[28] ferrous iron,[94-97] and manganese.[94, 96]

Ozone is an oxidant that may be overlooked by many because of its relative ineffectiveness in conventional waste treatment. It must be borne in mind that a third stage of sewage treatment will start with an effluent that is relatively low in organic matter and in other impurities. In such circumstances the high oxidizing power of ozone may operate particularly well. Furthermore, new methods of producing ozone will doubtless reduce the cost of ozonization by a considerable amount. The possible value of ozonization as a third-stage treatment method obviously has not yet been recognized, as is evident from the lack of research in this area. There is little discussion[98, 99] on ozonization in the literature. A recent paper by Nebolsine[69] affords some comparison of ozone with chlorine and chlorine dioxide for the oxidation of phenols in steel plant wastes.

Aside from its recognized value as a bactericidal agent - papers on this topic are abundant in the literature[100-105] - elemental chlorine has had limited use as an oxidant in waste water treatment. Divalent manganese[71] may be oxidized with chlorine, as may ferrous iron[71, 97] and cyanide.[72]

Many organoleptic substances in sewage effluents may be oxidized by chlorine, ozone, or chlorine dioxide. Neutral organic materials and organic acids have been oxidized with chlorine,[106-108] thereby reducing tastes and odors. Oxidation of phenol and amino acids with chlorine has been accomplished; specific instances and values may be found in the literature for valine, aspartic acid, glycine, alanine, tryosine, and other organic substances.[109]

Chlorine dioxide has also been used with fair success for oxidation of some of the above-mentioned waste constituents.[43, 69] It falls into a category somewhat similar to ozone in that it is an extremely strong oxidant and may "come into its own," so to speak, in application to effluents with relatively low concentrations of impurities.

On the whole, oxidation methods using oxidants of these types are limited by the selectivity of the oxidation processes; that is, some types of organic structures are readily oxidized, whereas others are not measurably attacked. Unless catalysts can be found to make these oxidants less discriminating, it is unlikely that they can serve as reagents for general solution of the problem of third-stage treatment.

The three methods of treatment that have been discussed offer the most nearly immediate possibilities for use in third-stage treatment of wastes. Other methods that have been proffered have been explored with respect only to very specific types of impurities. Instead of examining each of these methods separately, it is more convenient to consider them in connection with the purifying techniques tested on individual impurities in sewage effluents. The most prominent of the organic impurities that have received attention are phenols, other taste- and odor-producing compounds, and synthetic detergents.

In many instances other methods for the removal of the impurity under consideration will have been previously discussed in connection with one or more of the first three treatment methods. The following sections deal only with those processes not previously covered.

Phenolic Compounds

Two important methods for treating phenols in waste waters remain to be considered. The first of these is biological degradation. Several good general discussions of this technique are available in the literature.[69, 110] Reid et al.[111] have claimed that biological slimes grown in drums removed phenols more slowly than those grown in aeration tanks, but that the former are tolerant to much higher concentrations of phenols than the latter. Obviously this technique is not suitable for further removal of materials that have already shown themselves resistant to biological oxidation in second-stage treatment.

Extraction methods for removal of phenols constitute the second process considered worthy of mention. Benzene has been used for the extraction of phenolic compounds from gas works waste waters; treatment of the benzene extract with sodium hydroxide has resulted in complete recovery of the phenol.[112] Extraction of phenol from domestic sewage has been successfully accomplished with branched ketones[113] and with various aromatic hydrocarbon solvents in the pH 6.5 to pH 9.5 range.[114] Industrial experience has shown that butyl acetate is an excellent extracting solvent for phenolic compounds.[115]

Commercial dephenolization of ammoniacal liquors by use of centrifugal extractors has been practiced on a 200,000-gallons-per-day basis.[116] This method does not appear to be economical for general application in waste water treatment, however, and standard extractive fluids will not efficiently remove other types of impurities. Some exploratory investigations of unusual extraction methods may be warranted.

Organoleptic Materials

The causative agents of odors and tastes in waters have been described in several papers,[117, 118] and their respective susceptibilities to biological degradation have been discussed in an article by Ettinger.[108] A vacuum degasification method has been described as successful in the removal of hydrogen sulfide and methane from waste waters.[119] The best removal of tastes and odors by this method was reported to have been achieved at low pressure, low pH, and low flow. This method has no general applicability.

Synthetic Detergents

A fairly comprehensive review of the current research program on detergents was presented at the 1959 annual meeting of the Ohio Sewage and Industrial Wastes Association.[120] Many investigators have been concerned with the effects of synthetic detergents on the efficiency of popular waste treatment methods.

McLaren[121] and McKinney[122] have presented comprehensive articles dealing with the nature of commercially used synthetic detergents, their chemical properties, and their effects on water and sewage treatment. The American Water Works Association classifies the general problems associated with the presence of synthetic detergents treatment as: (1) foaming in settling basins, (2) taste and odor problems, (3) hindering of coagulation and sedimentation, (4) foaming of the finished waters, and (5) quality deterioration in the distribution system.[43]

A study of 11 representative surface active agents of anionic and non-ionic types, along with the building com-

pounds in greatest general use in the compounding of detergents, showed that none of these agents interfere with coagulation in concentrations below 10 milligrams per liter.[91] A study conducted by Ingols[123] showed that in the presence of synthetic detergents the sedimentation process is much less effective, however. Aultman[124] has presented a discussion of the effects of synthetic detergents upon water softening processes.

Mose[125] has reported, as have McKinney and Symons,[126] that alkylbenzenesulfonate, in the concentrations now found in sewage and even in forseeable concentrations, does not adversely affect biological processes. The work of McKinney and Symons has indicated that the major portion of the alkylbenzenesulfonate passing through a treatment plant is degraded in the secondary or biological stage. This observation is not entirely in accord with other experiments or practical experience.

Frothing seems to be the worst problem posed by synthetic detergents in treatment plants. Polkowski et al.[127] are engaged in research on foaming problems and have recently published some of their findings. Taste and odor problems resulting from synthetic detergents are of importance when multiple reuse of waters occurs. Some work has already been done on the removal of trace quantities of alkylbenzenesulfonate from water supplies. Renn and Barada[128] recently reported results of studies in which various particulate materials were used as adsorbents. Their results indicate that about 40 milligrams per liter of the commonly used types of adsorbing carbons are required to remove 90 percent of an original concentration of alkylbenzenesulfonate equal to 0.2 milligram per liter. When the alkylbenzenesulfonate was present at an initial concentration of 2 milligrams per liter, 95 milligrams per liter activated carbon was required to achieve 90 percent removal.

Many unusual techniques have been investigated for reduction of the concentration of alkylbenzenesulfonate in effluents from sewage plants. Investigators at the Sanitary Engineering Research Laboratory of the University of California have studied foam formation and surface burning as well as many other standard methods. Such techniques, which make use of special properties of synthetic detergents, may be useful in instances where the presence of such detergents is the only major problem, but are not capable of general application to all types of pollutants. The major reason for the growing concern about synthetic detergents is that many of them are not biologically degradable, or are only partially degradable. With the ever-increasing use of detergents, it is apparent that problems with them are continually becoming more pronounced. One solution that is currently under very active study is the possibility of substituting detergent materials that are sub-

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ject to biological degradation. For example, sugar-based detergents, which have been studied by investigators at the Water Pollution Research Laboratory of Great Britain,[42] have been found to be readily oxidized.

Phosphate and Nitrate

The problem of the efficient removal of the fertilizing ions, phosphate, and nitrate from sewage has long been under consideration; since these are essential nutrients for most aquatic flora, their presence in receiving water may result in troublesome algal blooms.

Precipitation of phosphate with aluminum, copper, and iron salts has given 99 percent reduction of phosphate in laboratory experiments.[85] The optimum pH range was found to be roughly between pH 7 and 8, with a conventional alum coagulant. Polyphosphate may be carried down with great efficiency at certain pH values, with aluminum hydroxide, ferric phosphate, or ferrous hydroxide; ferrous sulfate can be used to good advantage in the removal of orthophosphate.[86]

A modified activated sludge method has been used for the removal of nitrate nitrogen from the wastes from certain AEC sites.[129] The wastes contained nitrate in excess of the permissible limit of 10 milligrams N per liter and could not be treated by conventional biological methods.

Imhoff suggested the possibility of using either spray irrigation over large areas or shallow ponds from which the floating aquatic plants and sediments are periodically removed[130] as techniques for utilization of nitrate and thus its removal from water. However, both methods are extremely expensive and require large areas. Spray irrigation has the obvious additional disadvantage of water loss.

By recirculating highly oxidized sewage effluent into the aeration tank of an activated sludge plant, Wuhrmann was able to produce denitrification by reduction of nitrate and oxidation of ammonium ion to nitrogen gas, thus obtaining marked removals of total nitrogen.[172]

Salinity

There is no point in belaboring the need for some process by which saline or brackish waters can be desalted to provide potable water supplies. Throughout the world valuable water is lying unused, because of its high salt content.

> "Water, water, everywhere,
> Nor any drop to drink."

The quoted couplet from Coleridge's "The Ancient Mariner"

well summarizes the pitiful plight of many regions of the earth. With an economical method to desalinize this water, vast arid lands could be transformed into veritable gardens, industries could spring to life, and many peoples of the world would find a new mode of life available to them.

The waters of the seas could doubtless supply the water needs of man for generations hence if properly treated. But even in our own inland watercourses the problem of salinity is increasing proportionately to the increase of water reuse, since each time a water is used the salt content is increased. This situation can reasonably be extrapolated to a condition in the not distant future when all waters of the country will be brackish, unless some desalinization treatment is enacted upon the effluents of the waste treatment plants before they are discharged into receiving bodies. The first demineralization plant for a U.S. municipal supply has already been installed at Coalinga, California.[131] This small system treats only 28,000 gpd, and the cost is extremely high (estimated at $1.00 per 1000 gallons), but it does indicate a trend in treatment practices.

Four methods for the desalting of waters are now popular. Evaporation is of course an age-honored device, as is the freezing technique. Relatively new methods include ion exchange and electrodialysis. A recent report was given on saline water conversion research being carried on in various countries, including information on papers presented at the symposium conducted by the Office of Saline Water of the U.S. Dept. of Interior and the National Research Council of the National Academy of Sciences.[132] Methods discussed in this paper include distillation, membrane processes, solar distillation, and freezing processes. A similar report was given by Jenkins, who discussed and outlined several other physical and chemical methods of separating salt from water.[133] The reader may also find a comparison of demineralization and evaporation methods in a discussion by Baron and Gabel[134] of the relative merits of the two systems for removing boiler water impurities. Cost advantages of mixed-bed demineralization methods over evaporation techniques were shown by the latter authors. Similar discussions by Smith and Richeimer,[135] Copeland,[136] and others may be found in the literature.[137-140]

Ion-exchange demineralizers have been discussed by Calise,[141] who describes current practices in multibed and mixed-bed exchangers. Calise has pointed out advantages of demineralizers over evaporators, and has presented a tabulation of the types of ion-exchange equipment in use. Ionics Incorporated has introduced an apparatus developed for the demineralization of brackish waters containing 1000 to 6000 milligrams of total dissolved solids per liter, and has advanced claims that the apparatus is much more economical than distillation.[142] A more recent comparison of

demineralization and distillation by Kemmer[143] includes a convenient table indicating which waste constituents are best removed by the various commercial demineralizing units.

Several years ago Eliasson[144] and Kressman[145] presented discussions of the basic mechanics of electrodialysis techniques for desalting waters, and more recently Mayers and Volckman[146] described some experiments carried out between 1953 and 1957 on the demineralization of mine waters by use of electrodialysis. The latter paper includes a discussion of the different types of permselective membranes and the advantage they offer over other types.

Distillation procedures are still the most widely used, and certainly the most widely discussed of the desalting techniques. The literature is crowded with papers describing various distillation installations and proposing many modifications and variations on this age-old desalting method. Many of these references are cited in the bibliography of this report and may be of some interest, although none seem to throw any bright new light on the subject.[147-156]

A unique compresion still has been devised for the pressure distillation of saline waters.[157] The still contains a rapidly rotating, sheet-metal, evaporator-condensor. Under centrifugal force liquid films on both sides of the rotor are maintained so thin that over-all heat-transfer coefficients of 2500 to 6000 Btu/ft^2/hr/°F are routinely developed. The paper has reported that tons of seawater have been distilled with a pair of 18-inch-diameter copper rotors without the development of scale on the rotors, and that a temperature differential as low as 3°F has been realizable.

Stanley[158] recently reported on the economic considerations of saline water conversion using solar distillation. In the research project report he presented a review of the literature on solar distillation of saline waters, and gave cost estimates for greenhouse-type installations in which solar rays are transmitted through a glass roof over an evaporating basin, the water condensing on the inner surface of the roof and being collected in channels as it runs down the glass.

Freezing is another method that has been given considerable consideration as a desalting procedure. Thompson and Nelson[159] have presented the results of investigations designed to obtain quantitative information on the distribution of salts in ice formed by freezing brackish water, and to ascertain the relative salinity of the waters obtained upon melting of the ice. More recently Hendrikson[160] discussed seawater desalting tests with commercial and laboratory freezers, including methods for the separation of the salt from the frozen brine mixture and some cost data.

As the situation presently stands, the method most suitable for the desalting of a saline or brackish water, from the realistic viewpoint of economics, is very largely a function of the salinity content of the water. For seawater, with its high salinity, it appears that a distillation or freezing procedure is to be preferred. However, as a step for salt removal in the treatment of waste waters, methods based on electrodialysis or ion exchange appear preferable. The latter methods are more economical for the treatment of a water that is relatively low in salt content, but become prohibitively expensive as the salinity increases. There is then an economic equilibrium point that must be considered in the selection of any desalting process.

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GPO 810-934-5